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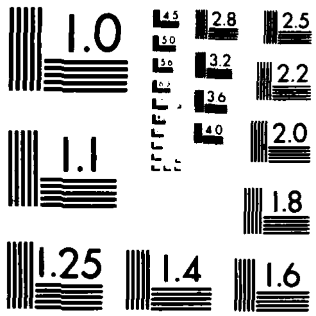

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TWO-STEP SEQUENTIAL REACTIONS FOR LARGE ACTIVATION ENERGIES-REVISITED

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ABSTRACT

One-dimensional, steady flame propagation for a sequential, two-step reaction of the form $A \xrightarrow{\text{yields}} B \rightarrow C$ is considered. An earlier investigation of the problem ~~by Kap~~ila and Ludford (Combustion and Flame <u>29</u>, p. 167 (1977)) determines that two separated flames generally exist and that their ordering is fixed by the ordering of the (disparate) magnitudes of the activation energies. The present work shows to the contrary that reversals of the flame ordering are quite possible, but that this is a subtle effect requiring attention to issues which are usually ignored in the theory of single flames.

AMS (MOS) Subject Classifications: 80A25, 34E05

Key Words: two-step sequential reaction, large activation energies, matched asymptotic expansion

Work Unit Number 2 (Physical Mathematics)

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SIGNIFICANCE AND EXPLANATION

Asymptotic methods based on large activation energies have been established over the last decade as an effective technique for analyzing chemical processes in combustion which are described by Arrhenius kinetics. However, efforts have been largely restricted to simple single-step reactions, which are of limited practical interest. The need for a more thorough treatment of multiple-step reactions has been recognized for some time and motivates the present investigation as a step towards narrowing the gap between the basic theory and the practical needs of engineering applications.

The contribution of this report to the analysis of multiple-step combustion is two-fold. It provides a careful treatment of the title problem and, more importantly, it demonstrates the sometimes subtle complexities introduced by the presence of more than one reaction. The need for attention to detail is exemplified as subtle differences in analysis are shown to lead to remarkably different conclusions.

The responsibility for the wording and views expressed in this descriptive summary lies with MRC, and not with the authors of this report.

TWO-STEP SEQUENTIAL REACTIONS FOR LARGE ACTIVATION ENERGIES-REVISITED

H. V. McConnaughey and G. S. S. Ludford

INTRODUCTION

This discussion is concerned with steady, one-dimensional propagation of the two laminar flames associated with the two-step sequential reaction $A \to B \to C$. Of particular interest are the admissible flame configurations and associated burning rates.

Kapila and Ludford [1] investigated this problem using an asymptotic analysis based on the limit of large activation energies, E_1 and E_2. It was found that two separated flames generally exist, but that the flames can also merge. The order in which the separated flames occur was shown to be fixed by the relative sizes of the activation energies. In all cases, the burning rate was determined explicitly.

This report presents a generalized version of [1] which also considers the limits $E_1 \to \infty$ and $E_2 \to \infty$. The generalization is subtle, however, involving only terms which are $O(E_1^{-1})$ or $O(E_2^{-1})$, and is likely to appear superfluous at first glance. Indeed, it reproduces the results found previously for the case of separated flames and yields burning-rate formulas for merged flames which are nearly identical to those in [1]. Nevertheless, the restriction on the separated-flame ordering found by Kapila and Ludford is eliminated.

The nature and significance of this generalization are best seen in the context of the asymptotics, which are therefore included in this discussion to the extent necessary. Treatment of separated flames is omitted since it does

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not appreciably modify the conclusions derived by Kapila and Ludford from their investigation of separated flames (see [2]). An analysis of merged flames is needed, however, and hence is presented here along with pertinent results for separated flames. The approach in [1] is followed, thus the reader is referred to that work for any necessary clarification.

It should be emphasized that most of the present analysis follows that of Kapila and Ludford, although extra terms may appear and the notation may differ slightly. The significant deviations from their work are explicitly stated.

The Mathematical Problem

Steady plane flame propagation for a sequential two-step reaction $A \to B \to C$ is considered. The governing dimensionless equations are

$$L(Y) = W_1' \ , \tag{1}$$

$$L(Z) = -W_1' + W_2' \ , \tag{2}$$

$$L(T) = -Q_1 W_1' - Q_2 W_2' \ , \tag{3}$$

where

$$L = \frac{d^2}{dx^2} - \frac{d}{dx} \ , \quad x \in (-\infty, \infty) \ ,$$

$$W_1' = \frac{D_1'}{M^2} Y \exp\left[\frac{E_1(T - T_1)}{TT_1}\right] \ ,$$

$$W_2' = \frac{D_2'}{M^2} Z \exp\left[\frac{E_2(T - T_2)}{TT_2}\right] \ ,$$

and where

Y = mass fraction of species A,

Z = mass fraction of species B,

T = dimensionless temperature,

subscript 1 ~ pertaining to reaction $A \to B$,

subscript 2 ~ pertaining to reaction $B \to C$,

$D_i' \exp(E_i/T_i) = D_i$ = Damköhler number,

$D_i' \propto E_i^{n_i}$, where n_i = unspecified constant,

E_i = dimensionless activation energy,

T_i = parameter characterizing magnitude of D_i,

M = burning rate,

Q_i = dimensionless heat release, $Q_1 + Q_2 = 1$.

(The reader is referred to [1] for additional details.) The boundary conditions are

$$x \to -\infty: \quad Y \to Y_{-\infty}, \quad Z \to Z_{-\infty}, \quad T \to T_{-\infty} ; \tag{4}$$

$$x \to +\infty: \quad Y \to 0, \quad Z \to 0, \quad T \to T_{\infty} . \tag{5}$$

Note that this system is invariant under translations, therefore the origin may be fixed as desired. Also, one of equations (1)-(3) may be eliminated by integrating the combination $(1) + Q_2(2) + (3)$ subject to (4) and (5). The resulting identity:

$$Y(x) + Q_2 Z(x) + T(x) \equiv Y_{-\infty} + Q_2 Z_{-\infty} + T_{-\infty} = T_{\infty} \tag{6}$$

holds.

The objective of Kapila and Ludford's investigation is to determine from (1)-(6) the variation of M with D_1 and D_2, the other parameters being fixed. This is accomplished by an asymptotic analysis based on large activation energies. In the limits $E_1 \to \infty$ and $E_2 \to \infty$, the chemical activity, represented by the nonlinear terms in (1)-(3), is localized in thin zones (flames) where the temperature is close to T_1 or T_2. Outside of these zones, the linear reactionless form of equations (1)-(3) holds. The resulting "outer" solutions are matched at the flames to the solutions of the (nonlinear) equations valid inside the reaction zones.

Two types of flames are possible: 1) separated flames, corresponding to cases where $T_1 - T_2 = O(1)$, by which the combustion field is divided into three reactionless regions, and 2) merged flames, which separate two chemically inert regions and occur when $T_1 - T_2 = O(E_j^{-1})$, $E_j = \min(E_1, E_2)$.

For convenience, we introduce the constants $\tilde{T}_1$ and $\tilde{T}_2$, where

$$\tilde{T}_i = \lim_{E_1, E_2 \to \infty} T_i \ .$$

In this notation, separated flames exist when $\tilde{T}_1 \neq \tilde{T}_2$; merged flames occur if $\tilde{T}_1 = \tilde{T}_2$.

Kapila and Ludford do not distinguish between T_k and $\tilde{T}_k$ when k is the index associated with the reaction which exhausts all reactants (i.e. beyond which $Y = Z = 0$). For that index, $\tilde{T}_k = T_\infty$ and in [1], it is effectively assumed that T_k is strictly equal to T_∞. It is this assumption which leads to the aforementioned restriction on the ordering of separated flames, as will be shown.

<u>Results for Separated Flames</u> $(\tilde{T}_1 \neq \tilde{T}_2)$

The reaction occurring at the lower temperature precedes (in location) the other reaction. For $\tilde{T}_1 < \tilde{T}_2$, the flames then appear as shown in Figure 1 and $\tilde{T}_2$ equals T_∞. The burning rate for this case obtained in [1] satisfies

$$M^2 = \frac{2T_\infty^4 D_2}{[(Y_{-\infty} + Z_{-\infty})E_2 Q_2]^2} \exp(-E_2/T_\infty) \ . \tag{7}$$

For $\tilde{T}_2 < \tilde{T}_1$, the flames in Figure 1 are reversed, $\tilde{T}_1 = T_\infty$, and M is given by

$$M^2 = \frac{2T_\infty^4 D_1}{(Y_{-\infty} E_1)^2} \exp(-E_1/T_\infty) \ . \tag{8}$$

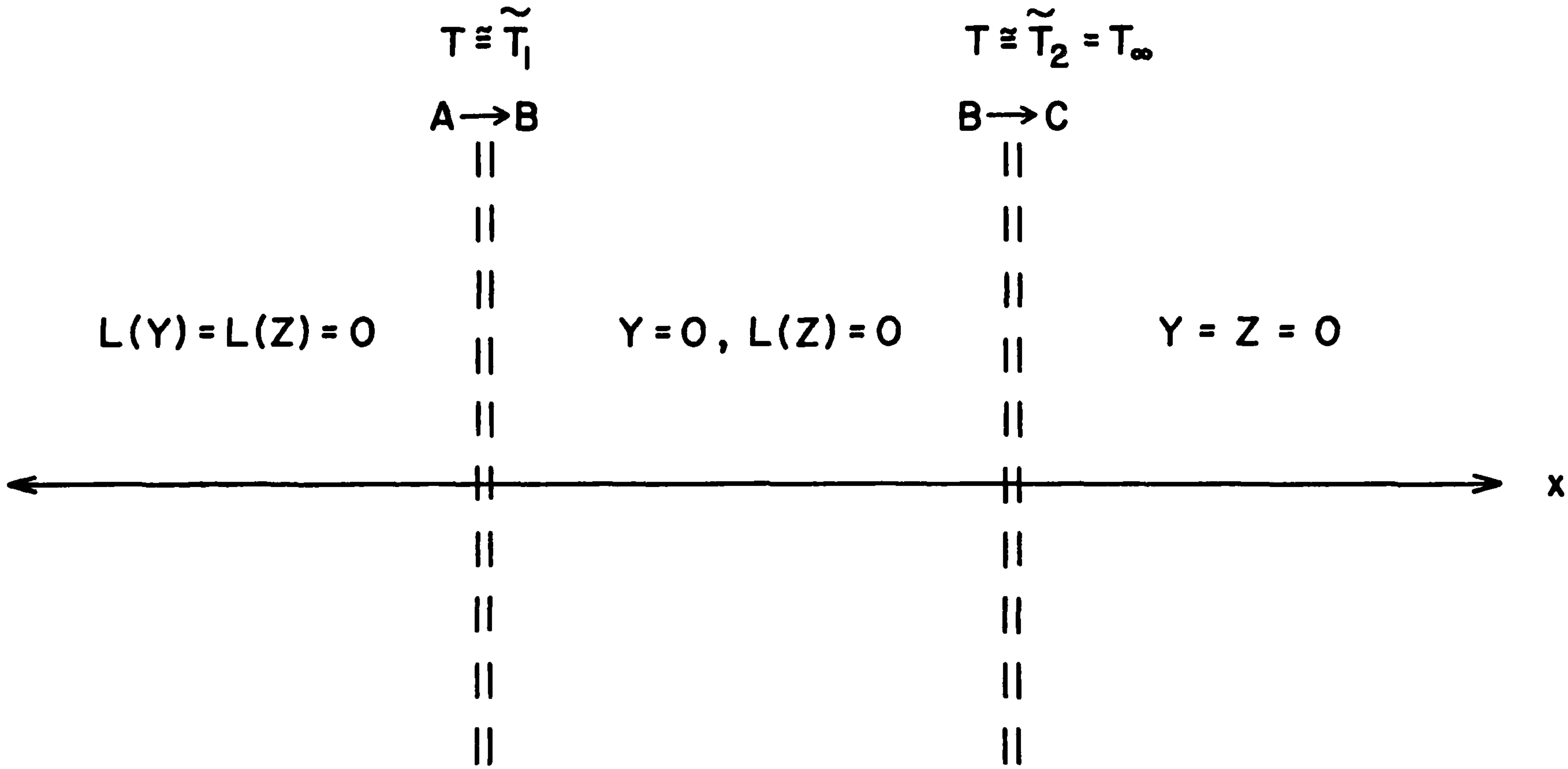

Figure 1. Separated flames for $\tilde{T}_1 < \tilde{T}_2$.

Merged Flames ($\tilde{T}_1 = \tilde{T}_2$)

When the flames are merged, the combustion field is divided into only two chemically inert regions, as represented in Figure 2a. The origin has been located within the flame zone. The solution of the reactionless equations for $x \neq 0$ subject to (4) and (5) is

$$x < 0: \quad Y = (1 - e^x)Y_{-\infty} + \nu e^x, \quad Z = (1 - e^x)Z_{-\infty} - \nu e^x, \quad T = T_\infty - Y - \rho_2 Z ,$$

$$x > 0: \quad Y = 0, \quad Z = 0, \quad T = T_\infty ,$$

where ν is an unspecified quantity of order E_j^{-1}. Continuity of Y and Z to leading order across $x = 0$ has been imposed; the origin has been fixed so that $Y + Z$ is independent of E_1 and E_2. The leading-order flame temperature $\tilde{T}_1 = \tilde{T}_2$ must equal T_∞.

The validity of the above outer solution is contingent upon the existence of a flame-zone solution or "structure" in the neighborhood of $x = 0$ which effects the change in slope there. The structure analysis below is seen to fix the burning rate M.

Equations (1) and (2) indicate that the $A + B$ reaction and the $B + C$ reaction become active when $T - T_i = O(E_i^{-1})$, $i = 1,2$. The small parameters δ_1 and δ_2 which gauge the thickness of the flames are thus chosen so that $\delta_i = O(E_i^{-1})$. Although the two reactions occur at the same $O(1)$ location $(x = 0)$, they must be distinct in order to make the structure problem analytically tractable. It is therefore assumed that the reaction zone thicknesses δ_1 and δ_2 are of disparate orders, or equivalently, the activation energies are of disparate orders.

Consider the case where $E_1 >> E_2$, so that $\delta_1 << \delta_2$. The associated merged-flame configuration is illustrated in Figure 2b. On the scale of the (broader) $B + C$ reaction, the $A + B$ reaction is located at $x/\delta_2 = \rho_0$ where it appears as a discontinuity. All chemical activity of the $A + B$

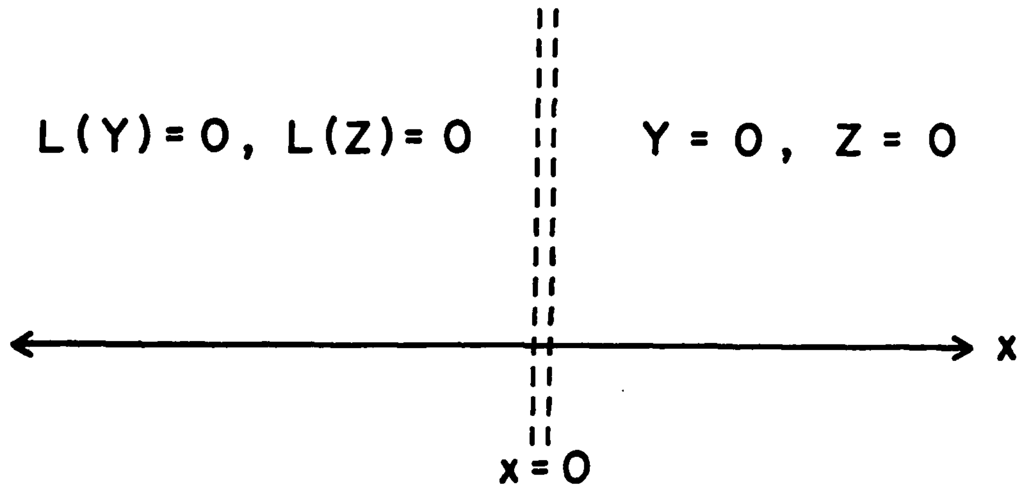

Figure 2a. Appearance of merged flames on the O(1) scale: A single discontinuity at x = 0.

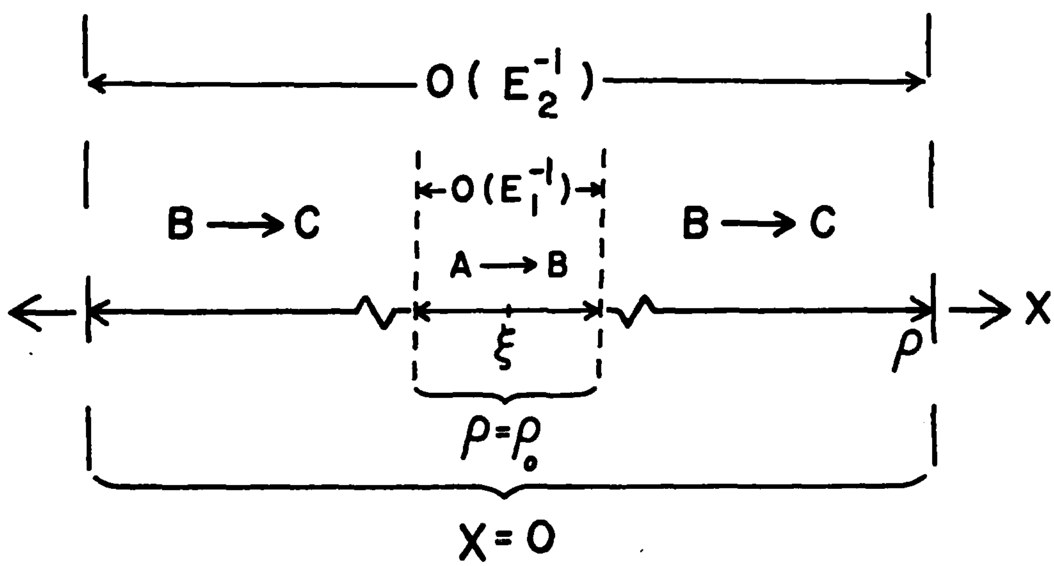

Figure 2b. Merged-flame configuration for $E_1 >> E_2$: A → B reaction zone appears as a discontinuity at ρ_0 on the scale of the broader B → C reaction.

reaction is localized and completed there; the reaction is frozen for $x/\delta_2 = \rho < \rho_0$ and is in equilibrium (reactant A is exhausted) for $\rho > \rho_0$. The $B \to C$ reaction, on the other hand, is active for $\rho \in (-\infty,\infty)$. It is only partially completed for $\rho < \rho_0$ and is not significantly affected at ρ_0 by the $A \to B$ reaction there since the latter occurs in such a thin zone. The amount of reactant B remaining at $\rho = \rho_0^-$, and all B produced at ρ_0, are completely converted into C over the interval (ρ_0,∞) of ρ.

Thick-Flame Structure

In the $B \to C$ reaction zone, the variables are written:

$$x = \delta_2\rho, \quad \rho \in (-\infty,\infty) ,$$

$$T = T_\infty + \delta_2 t(\rho) + o(\delta_2) ,$$

$$Z = \delta_2 z(\rho) + o(\delta_2) ,$$

$$Y = \begin{cases} \delta_2 y(\rho) + o(\delta_2) & \text{for } \rho < \rho_0 \\ 0 & \text{for } \rho > \rho_0 , \end{cases}$$

where $\delta_2 = T_\infty^2/E_2$, $t(\rho) = -y(\rho) - Q_2 z(\rho)$ for $\rho < \rho_0$ and $t(\rho) = -Q_2 z(\rho)$ for $\rho > \rho_0$. The parameters T_1 and T_2 may similarly be expressed as

$$T_1 = T_\infty + \delta_2 t_1 + o(\delta_2), \quad T_2 = T_\infty + \delta_2 t_2 + o(\delta_2) .$$

(Kapila and Ludford assume $t_2 = 0$.) To leading order, equations (1) and (2) are reduced to

$$\rho < \rho_0 : \frac{d^2y}{d\rho^2} = \delta_2^2 D_1' M^{-2} y(\rho)\exp\{[t(\rho) - t_1]E_1/E_2\} , \tag{9}$$

$$\frac{d^2z}{d\rho^2} = -\frac{d^2y}{d\rho^2} + \delta_2^2 D_2' M^{-2} z(\rho)\exp[t(\rho) - t_2] ,$$

and

$$\rho > \rho_0 : Y = 0, \quad \frac{d^2z}{d\rho^2} = \delta_2^2 D_2' M^{-2} z(\rho)\exp[t(\rho) - t_2] .$$

Note that since $E_1/E_2 >> 1$, $t(\rho) - t_1$ must be negative for $\rho < \rho_0$ in order that equation (9) reflect a frozen $A \to B$ reaction. Impose then

$$t(\rho) < t_1 \quad \text{for} \quad \rho < \rho_0 , \tag{10}$$

and define the $O(1)$ constant $\tilde{D}_2$ as $\tilde{D}_2 = \delta_2^2 D_2'$ (hence $n_2 = 2$). The $B \to C$ structure equations then become

$$\rho < \rho_0 : \frac{d^2 y}{d\rho^2} = 0, \quad \frac{d^2 z}{d\rho^2} = \tilde{D}_2 M^{-2} z \exp(-y - Q_2 z - t_2), \quad \text{and}$$

$$\rho > \rho_0 : Y = 0, \quad \frac{d^2 z}{d\rho^2} = \tilde{D}_2 M^{-2} z \exp(-Q_2 z - t_2) .$$

Matching with the outer solution provides the conditions

$$\rho \to -\infty : \frac{dy}{d\rho} \to -Y_{-\infty}, \quad \frac{dz}{d\rho} \to -Z_{-\infty}, \quad \text{and}$$

$$\rho \to +\infty : \frac{dz}{d\rho} \to 0, \quad z \to 0 .$$

Also y, z, and $d(y + z)/d\rho$ must be continuous at ρ_0. It follows that

$$y(\rho) = (\rho_0 - \rho) Y_{-\infty} \quad \text{for} \quad \rho < \rho_0 ,$$

$$z(\rho_0^-) = z(\rho_0^+) = z(\rho_0) , \quad \text{and}$$

$$\frac{dz}{d\rho}(\rho_0^-) = \frac{dz}{d\rho}(\rho_0^+) + Y_{-\infty} .$$

The value of ρ_0 is not yet known, however. It must be fixed at the point where the $A \to B$ reaction becomes important, i.e. where $T - T_1 = O(E_1^{-1})$. Thus, $t(\rho_0) - t_1$ must vanish. This fact guarantees (10) since $d^2t/d\rho^2 < 0$ and $dt/d\rho$ is seen to be positive at ρ_0^-. It also gives $Q_2 z(\rho_0) = -t_1$. Since z is the leading-order term in Z, $z(\rho_0)$ must be positive, requiring t_1 to be negative, which is not surprising. A

negative t_1 requires that T_1 remain less than T_∞ through $O(\delta_2)$, ensuring that the $A \to B$ reaction remain frozen throughout the region preceding its zone, as is necessary.

The $B \to C$ structure problem may now be written:

$$\left.\begin{aligned}
&\frac{d^2z}{d\rho^2} = \begin{cases} \tilde{D}_2 M^{-2} z \exp[(\rho - \rho_0)Y_{-\infty} - Q_2 z - t_2] & \text{for } \rho < \rho_0 \\ \tilde{D}_2 M^{-2} z \exp(-Q_2 z - t_2) & \text{for } \rho > \rho_0 \,, \end{cases} \\
&z \to Z_{-\infty}\rho \to -Y_{-\infty}\rho_0 \quad \text{as} \quad \rho \to -\infty \,, \\
&z(\rho_0) = -Q_2^{-1} t_1 \,, \\
&\frac{dz}{d\rho}(\rho_0^-) = \frac{dz}{d\rho}(\rho_0^+) + Y_{-\infty} \,, \\
&\frac{dz}{d\rho} \to 0, \quad z \to 0 \quad \text{as} \quad \rho \to +\infty \,.
\end{aligned}\right] \tag{11}$$

If $\tilde{D}_2\exp(-t_2)$ is labeled $\bar{D}_2$ and t_1 is labeled $-t_0$, (11) takes on the form of the $B \to C$ structure problem obtained and numerically solved in [1]. Those numerical results give the equivalent of

$$\begin{aligned} M^2 &= \tilde{D}_2\exp(-t_2)/F(-t_1) \\ &= E_2^{-2} T_\infty^4 D_2 \exp(-E_2/T_\infty)/F(-t_1) \,, \end{aligned} \tag{12}$$

where F exhibits the asymptotic behavior

$$F \sim \begin{cases} \frac{1}{2} Q_2^2 (Y_{-\infty} + Z_{-\infty})^2 & \text{as} \quad t_1 \to -\infty \\ Q_2^2 Y_{-\infty}^2 / t_1^2 & \text{as} \quad t_1 \to 0 \end{cases} \tag{13}$$

and is illustrated in the graph in Figure 3 for $Q_2 = .5$ and $Y_{-\infty} = .75$ (Ref. [1] includes an extra factor of $\frac{1}{4}$ in the behavior of F as $t_1 \to 0$).

In the limit $t_1 \to -\infty$, $\tilde{T}_1$ no longer equals T_∞ but rather $\tilde{T}_1 < T_\infty$, which implies a separated-flame configuration with the $A + B$ reaction preceding the $B + C$ reaction. Result (12) should therefore yield result (7) in the limit $t_1 \to -\infty$, which it does according to (13). Also, problem (11) requires that $z(\rho_0) \to \infty$ as $t_1 \to -\infty$, thus implying that $\rho_0 \to -\infty$ or equivalently that the $A + B$ reaction zone moves to the left of the $B + C$ flame.

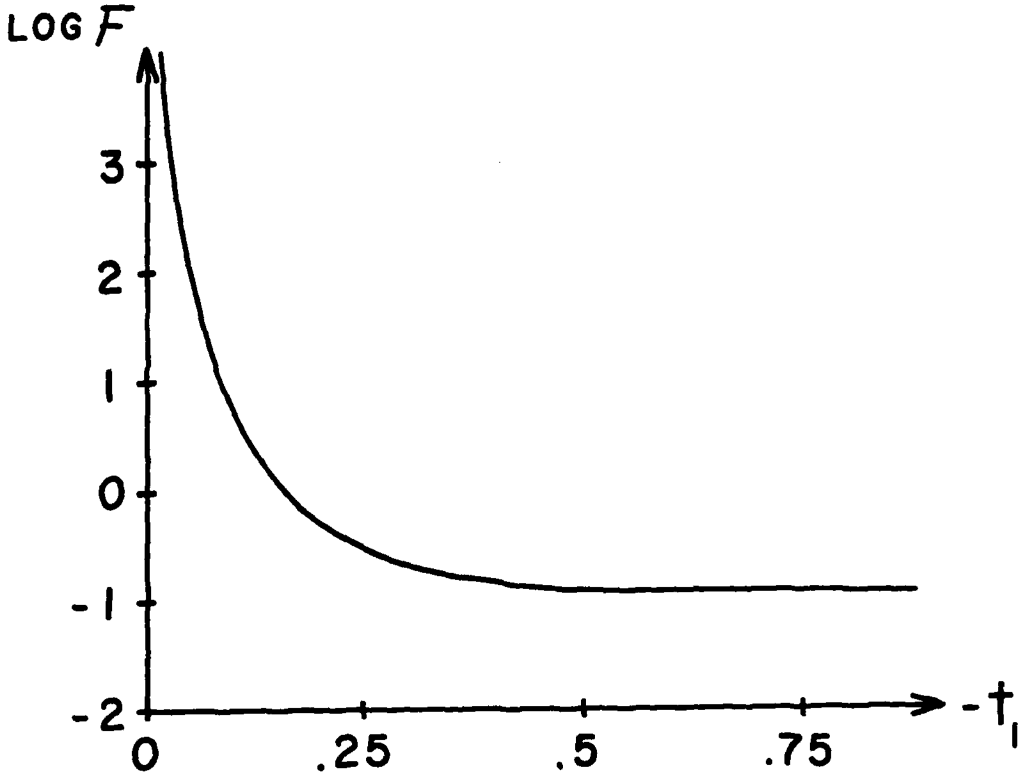

Figure 3. Graph of $F(-t_1)$ for $\rho_2 = .5$ and $Y_{-\infty} = .75$ as calculated by Kapila and Ludford [1].

The limit $t_1 \to 0$, on the other hand, gives $T_1 = T_\infty + o(\delta_2)$ and may also represent separated flames if $T_\infty - T_2$ becomes $O(1)$. (Separated flames occurring for $\tilde{T}_2 < \tilde{T}_1$ correspond to $0 < T_\infty - T_2 = O(1)$ and $T_1 = T_\infty + O(\delta_1)$.) It is therefore possible that the corresponding burning

rate (8) may be recovered from (12) and (13) if the proper distinguished limits describing $t_1 \to 0$ and $t_2 \to -\infty$ are considered. This requires a careful asymptotic analysis, however, and is not pursued here. It may be noted, though, that the limit $t_1 \to 0$ gives $z(\rho_0) \to 0$ in (11), implying completion of the $B \to C$ reaction, that is $\rho_0 \to \infty$. This indicates that the $A \to B$ flame moves to the right of the $B \to C$ flame.

It now appears feasible that either of the two possible separated-flame configurations may be recovered from the merged-flame configuration just considered. It follows that it may be possible for separated flames to merge and subsequently separate with their ordering reversed; i.e. the flames may cross as T_1 and T_2 are varied. This conclusion contradicts that of Kapila and Ludford.

Although the result (12) has the identical form of the burning rate found in [1], there is one crucial difference between the two results: the nonnegative argument of F. In the present work, the argument of F is

$$-t_1 = \delta_2^{-1}(T_\infty - T_1)$$

and is naturally nonnegative. The argument of F in [1], on the other hand, is

$$t_0 = \delta_2^{-1}(T_2 - T_1) ,$$

so that the restriction $t_0 > 0$ leads to the conclusion that $T_1 < T_2$ when $E_1 >> E_2$, admitting only one type of separated flames. Indeed, if T_1 cannot exceed T_2, the separated flame configuration represented by (8) cannot exist, hence the flames cannot cross. It follows that the ordering of separated flames is fixed by the ordering $E_1 >> E_2$. The source of this restrictive conclusion is Kapila and Ludford's unspoken assumption that T_2 exactly equals T_∞, i.e. that $t_2 = 0$.

The structure analysis for the thinner flame contributes only information about higher-order terms in the outer solution. Treatment of the inner structure problem is therefore omitted.

Analogous results and conclusions are obtained from the analysis of merged flames with $E_2 >> E_1$. In [1], it is found that T_2 cannot exceed T_1 in that case, hence only separated flames represented by (8) are possible. This follows from the assumption that T_1 is exactly T_∞. If a more general T_1 is allowed, i.e., $T_1 = T_\infty + \delta_1 \tau_1$, say, the restriction on admissible separated flames need no longer be true.

Conclusion

The present investigation, of which the analysis in [1] constitutes a special case, assumes general forms for the characteristic temperatures T_1 and T_2 by distinguishing T_i $(i = 1,2)$ from its limit at infinite activation energies. The burning-rate results of Kapila and Ludford are found to be true in general for separated flames, but must be modified for merged flames. Consequently, some of their conclusions are not generally applicable. In particular, their analysis of merged flames forces them to conclude that the order in which separated flames occur is dictated by the relative sizes of the activation energies. This restriction is a consequence of their assumption that one of the characteristic temperatures is strictly equal to its limit. By accounting for the difference, this work removes that restriction.

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2. McConnaughey, H. V., Three Topics in Combustion Theory, Ph.D. Thesis, Cornell University, pp. 86-103 (1983).

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20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

One-dimensional, steady flame propagation for a sequential, two-step reaction of the form $A \to B \to C$ is considered. An earlier investigation of the problem by Kapila and Ludford (Combustion and Flame 29, p. 167 (1977)) determines that two separated flames generally exist and that their ordering is fixed by the ordering of the (disparate) magnitudes of the activation energies. The present work shows to the contrary that reversals of the flame ordering are quite possible, but that this is a subtle effect requiring attention to issues which are usually ignored in the theory of single flames.

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